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On the Intensity Calculation of Multiple Reflexions of X-rays

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Expressions are derived for the set of structure factors, and for the polarization and Lorentz factors due to the multiple reflexion of X-rays.

Introduction

If there are more than two points of the reciprocal lattice (its origin included) sufficiently near to the reflexion sphere, the Bragg condition is fulfilled simultaneously for several directions of Bragg reflexion (simultaneous reflexion). In this case every reflexion beam necessarily finds several possibilities of being reflected again and again (serial reflexion). Therefore simultaneous reflexion and serial reflexion are two sides of the same physical fact, multiple reflexion.

Frequently such multiple reflexions (in their simplest form known as double reflexion, Umweganregung, Renninger effect) are advantageously used for investigating the perfection of crystal surfaces and determining lattice parameters (Renninger, 1937; Cole, Chambers & Dunn, 1961; Melle, 1974). In some cases multiple reflexions complicate the interpretation of the intensity of reflexion (Renninger, 1960; Moon & Shull, 1963), for example in the calculation of structure factors. Therefore it is necessary to be quite clear about the set of structure factors which are involved in multiple reflexion. It is also necessary to know the polarization factor and the Lorentz factor for the registration technique mainly used (generalized equi-inclination technique).

The set of structure factors

There are p points of the reciprocal lattice sufficiently near to the reflexion sphere and determined by the vectors h_n $(n=0,1,2,\ldots p-1)$. These points of the reciprocal lattice are conjugated to the structure factors F_n. The intensities of the p reflexions determined by the wave vectors $\mathbf{K}_n$ ('first generation') are essentially influenced by the structure factors F_n. Each of these p reflexion beams may be considered as an incident beam (James, 1958). If this is the case for each index m, the reciprocal-lattice points described by the vectors $\mathbf{h}_{m-n}=\mathbf{h}_m-\mathbf{h}_n$ $(m=0,1,2,\ldots p-1)$ are necessarily on the reflexion sphere. Thus a new set of beams is produced ('second generation'), and also a new set of structure factors $F_{m-n}=F(\mathbf{h}_{m-n})$ influencing the intensity of these beams.

Because F_{m-n} and F_{n-m} are conjugate complex it is possible to arrange the structure factors of the 'first and second generation' in the form of a Hermitian matrix

$$\mathsf{F}=(F_{i-j})\quad (i,j=0,1,2\ldots p-1)\,. \tag{1}$$

The vectors of the reciprocal lattice involved are described by a skew symmetric matrix

$$\mathsf{h}=(\mathbf{h}_{i-j})\,. \tag{2}$$

New directions of beams are not generated because the vectors $\mathbf{h}_{m-n}$ are produced by a simple translation of the vectors $\mathbf{h}_n$. So the set of wave vectors can be arranged in a single-column matrix

$$\mathsf{K}=(K_i)\,. \tag{3}$$

In an analogous manner a 'third generation' of reflexions arises, with the vectors $\mathbf{h}_{l-n}-\mathbf{h}_{m-n}$ $(l=0,1\ldots p-1)$ participating. Now

$$\mathbf{h}_{l-n}-\mathbf{h}_{m-n}=\mathbf{h}_{l-m} \tag{4}$$

and the corresponding structure factors are already included in the 'first and second generation', but there are no new directions of the beams. In the 'first generation' the direction of K_n is conjugated to the structure factor F_n, but in the 'second generation' already the whole set of the structure factors F_{m-n} influences the intensity of the beam in the direction of K_n. Therefore all structure factors F_{m-n} are to be taken into account in the calculation of the intensity. How this is to be performed depends on the perfection of the crystal-lattice and the degree of coherence of the X-rays.

Polarization factors

The influence of the reflexion on unpolarized X-rays may be described by the matrix

$$\mathsf{P}_i=\begin{pmatrix}1 & 0\\ 0 & \cos 2\theta_{i-1,\,i}\end{pmatrix} \tag{5}$$

where $2\theta_{i-1,i}$ is the Bragg angle between the $(i-1)$th beam and the ith beam.

In order to calculate the polarization correction of further reflexions it is useful to consider the components of the optical field parallel and perpendicular to the planes of incidence (Fig. 1). For each of these components the polarization correction has to be determined. The calculations can be readily surveyed if matrices for the spatial rotation of the coordinate systems described by the planes of indicidence are used as well. By means of the transformation matrix

$$\mathsf{T}_i = \begin{pmatrix} \cos\psi_{i-1,i} & \sin\psi_{i-1,i} \\ -\sin\psi_{i-1,i} & \cos\psi_{i-1,i} \end{pmatrix} \quad (6)$$

we obtain after j reflexions the resulting polarization factor

$$P = \tfrac{1}{2}\mathsf{P}_1\mathsf{T}_1\mathsf{P}_2 \ldots \mathsf{T}_{j-1}\mathsf{P}_{j-1} \quad (7)$$

where $\psi_{i-1,i}$ is the angle between the normal of the $(i-1)$th plane of incidence and the ith one.

After performing the matrix multiplication (7) the matrix elements are squared and added in the case of a mosaic crystal. For two reflexions we get with Zachariasen (1965)

$$\cos\psi_{12} = \frac{\cos 2\theta_0 - \cos 2\theta_{01}\cos 2\theta_{12}}{\sin 2\theta_{01}\sin\theta_{12}} \quad (8)$$

the polarization correction for the mosaic crystal

$$P = \tfrac{1}{2}[\cos^2 2\theta_{01} + \cos^2 2\theta_{12} + (\cos 2\theta_0 - \cos 2\theta_{01}\cos 2\theta_{12})^2] \quad (9)$$

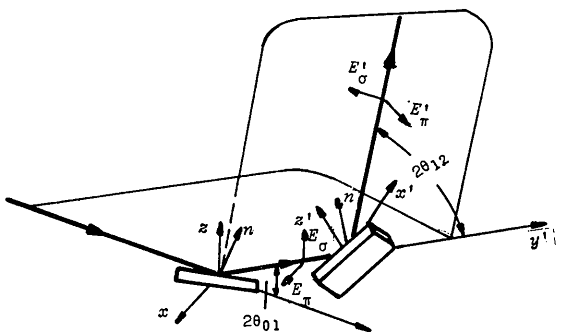

Fig. 1. Illustrating the nomenclature used in calculation of polarization factors. E_σ indicates the electric field component in the plane of incidence and E_π the component normal to it. The z axis is in the plane of indicidence and normal to the incident beam.

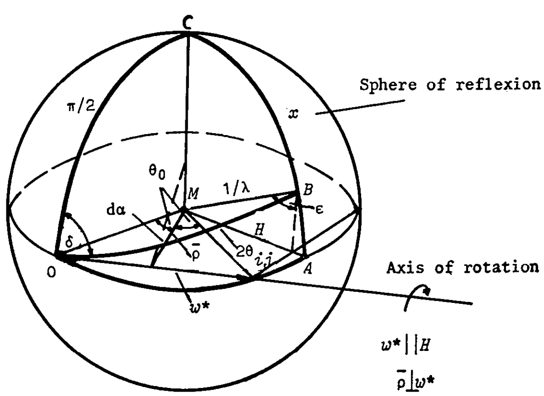

Fig. 2. Illustrating the nomenclature used in calculation of Lorentz factors.

in agreement with the result of Caticha–Ellis (1969), where $2\theta_0$ is the angle between the incident beam and the doubly diffracted beam.

Lorentz factor

For the recording of multiple reflexions it is convenient to use a generalized equi-inclination technique.

The generalized equi-inclination technique makes it possible to record reflexions represented by reciprocal-lattice points arranged in any plane of the reciprocal lattice perpendicular to the axis of rotation. We gain the generalized Lorentz factor by making use of the expression derived by Laue (1960) for the goniometer technique. Substituting the distance $\overline{MM}$ in Laue's figure by $\bar{\varrho}\,.\,\alpha$ with $\bar{\varrho} = 1/\lambda\cos\theta_0$ we obtain

$$L \sim \frac{1}{\bar{\varrho}\cos\chi} \sim \frac{1}{\cos\theta_0\cos\chi}\,. \quad (10)$$

By using the relations of the sperical trigonometry we obtain (triangle OBC, Fig. 2)

$$\cos\chi = \sin 2\theta_{ij}\cos\delta \quad (11)$$

and (triangle OAB)

$$\sin\varepsilon = -\sin\theta_0\cos 2\theta_{ij} + \cos\theta_0\sin 2\theta_{ij}\sin\delta \quad (12)$$

and

$$\cos\delta = \frac{\{\cos^2\theta_0\sin^2 2\theta_{ij} - (\sin\varepsilon + \sin\theta_0\cos 2\theta_{ij})^2\}^{1/2}}{\cos\theta_0\sin 2\theta_{ij}}\,. \quad (13)$$

With $\sin\varepsilon = \mathbf{H}\,.\,\lambda$ the Lorentz factor for the generalized equi-inclination technique may be written as

$$L_{ij} = \{\cos^2\theta_0\sin^2 2\theta_{ij} - (\sin\varepsilon + \sin\theta_0\cos 2\theta_{ij})^2\}^{-1/2}\,. \quad (14)$$

This expression also contains the special cases for the Bragg reflexion ($\varepsilon = 0$, $\theta_0 = 0$), the rotating crystal method ($\sin\varepsilon = 2\sin\varphi\sin\theta$), and equi-inclination technique, ($\varepsilon = \theta_0 = \mu$) where φ is the angle between the axis of rotation and the reflecting plane and $(\pi/2 - \mu)$ is the angle between the axis of rotation and the incident X-ray beam.

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